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EXPLOSIVE INTERACTION OF MOLTEN UO<sub>2</sub> AND LIQUID SODIUM

by

D. R. Armstrong, G. T. Goldfuss, and R. H. Gebner

**BASE TECHNOLOGY** 



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#### ABSTRACT

This interim report describes a continuation of the work reported in ANL-7890, Interaction of Sodium with Molten  ${\rm UO}_2$  and Stainless Steel Using a Dropping Mode of Contact. In the current study, sodium was injected into a pool of molten  ${\rm UO}_2$ . The experiment consistently produced vapor explosions, both with the injection nozzle above and beneath the surface of the  ${\rm UO}_2$ . Although the efficiency of the conversion of thermal to mechanical energy was small (due in part to very conservative data analysis and an inefficient geometry), the results did demonstrate that there is no intrinsic reason why reactor materials cannot produce a vapor explosion.

## I. INTRODUCTION

The work described in this report is a continuation of the experiments reported in ANL-7890. In this study small amounts of sodium (<5 g) were injected into a pool of molten  $UO_2$  (<100 g).

## II. APPARATUS

The experimental facility consisted of five principal systems: (1) an induction generator and coil for melting the  ${\rm UO}_2$ ; (2) a furnace for heating the sodium; (3) a vacuum chamber which contained the induction coil and furnace; (4) a mechanical-drive mechanism for effecting the sodium injection; and (5) an instrumentation system which included two high-speed cameras, a force transducer, and pressure transducers.

The experiments were carried out in a water-cooled, stainless steel furnace chamber (12 in. in diameter by 36 in.) equipped with quartz viewports and containing a system for injecting sodium into molten  $\mathrm{UO}_2$ , as shown schematically in Fig. 1. The sodium was contained in a stainless steel bellows (i.e., injection pump) that connected to the injection tube, initially located within a resistance-heated sodium furnace. When the  $\mathrm{UO}_2$  in an inductively heated tungsten crucible reached the desired temperature, the injection pump was driven downward so that the injection tube extended out of the furnace to a predetermined point, either above or below the surface of the  $\mathrm{UO}_2$ . A second mechanism, mechanically coupled to the insertion drive, moved away a

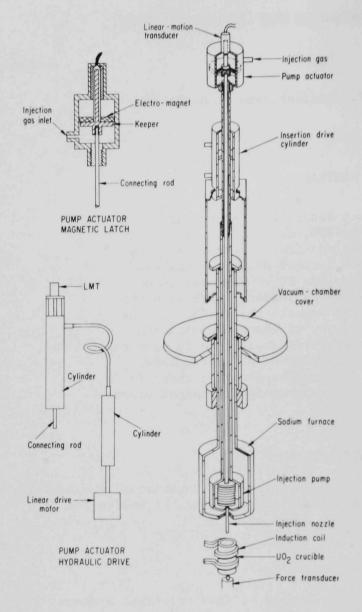


Fig. 1. Sodium Injector.
ANL Neg. No. 900-2556.

set of heat shields that covered the open end of the crucible during heating of the UO2. A simple counter-weighted flapper covered the opening in the bottom of the sodium furnace, shunting any sodium expelled from the injector, due to expansion during heating, into a trap.

Two techniques for injecting sodium into UO2 were used. In the first method, the bellows were pressurized and then released by deenergizing the matnetic latch. In the second method the bellows were released first and then pressurized by gas flowing through an orifice. A linear-motion transducer attached to the bellows monitored the injection rate.

The crucible containing the  $\mathrm{UO}_2$  was of wrought tungsten of 7/8-in. OD with a cavity 5/8 in. in diameter and 2 in. deep. It was supported by a tantalum rod holding a radiation shield and connected directly into a piezoelectric force transducer.

The crucible was inductively heated by a work coil made of 1/4-in.-sq copper tubing with a 16-gauge copper strip brazed onto the inner face of the coil as protection against arc penetration. The

work coil was silver-plated and silicon-oil-cooled. It was connected to an oil-cooled coaxial feedthrough. Power was supplied to the coil from a 100-kW, 450-kC generator feeding a 7:1 step-down transformer through a sheet-glass-insulated, parallel-plate transmission line.

The furnace chamber was normally operated in an environment of 1-atm of helium. A purge stream of helium was supplied to the main furnace chamber and exhausted from the sodium pump. Thus sodium vapor generated in the sodium furnace was swept out of the system.

A blast microphone for measuring the overpressure of the interaction was suspended above the crucible, and a high-speed motion-picture camera recorded the event at rates as high as 20,000 fps. Crucible temperatures were

measured by an optical pyrometer. Signals from the force transducer and blast microphone, along with timing signals applied coincidently to the cameras, were recorded on a 14-channel FM tape recorder.

The system has been operated at sodium temperatures ranging from the melting point to above  $600^{\circ}\text{C}$  and at  $\text{UO}_2$  temperatures up to  $3200^{\circ}\text{C}$ . Measured injection velocities ranged from 0.35 to 35 m/sec. The orifice-control pressurization provided constant injection velocities (nonlinearity <2%) over a range of 2 to 15 m/sec. There was a slight perturbation at the beginning of the injection due to the residual magnetic field in the latch, which required about 5 psi to overcome.

### III. PROCEDURE

The experimental data collected in this study consisted of: tank reaction force and gas-overpressure histories, high-speed motion pictures of the events, and the displacement of the sodium-injector bellows.

The sodium-injector assembly was weighed empty and placed in a helium glovebox. It was loaded with sodium by evacuating the bellows and backfilling with liquid sodium. The injection tube was capped with a slip-fit plastic cap, and the injector was removed from the glovebox, weighed, and assembled into the rest of the apparatus. A weighed quantity of crushed  $\rm UO_2$  pellets was added to the crucible, and all mechanical adjustments and alignments were made.

The furnace chamber was evacuated and filled with helium several times. A viewing port was opened and the cap on the injection tube removed. The chamber was evacuated and filled with helium several more times, and a helium purge started. At this time the sodium heaters were started. When the sodium reached the desired temperature, the induction generator was started and the  $\rm UO_2$  was heated at a rate of about  $100^{\circ}\rm C/min$ . When the selected temperature was reached and held for 2 to 5 min, an automatic sequencing apparatus was started. This turned on all of the recording apparatus at the proper times, and controlled the motion of the sodium injector and the injection of the sodium.

At the conclusion of each run, a reference signal was placed on the tape through each transducer amplifier system. The transducers were calibrated before each experiment.

## IV. RESULTS

A total of 12 experimental runs were made by injecting sodium into molten  $\mathrm{UO}_2$ . Nine of these were above-surface injections and three beneath the surface. Five of the 12 runs produced explosions. The criterion for an explosion was the production of a shock wave in the atmosphere surrounding the crucible, either as determined from the blast-microphone records or by the noise transmitted through the apparatus. Table I shows the initial conditions for all 12 experiments. In three (5, 10, 11) runs there were no explosions, as the sodium was prevented from contacting the  $\mathrm{UO}_2$  because of mechanical failures in the apparatus. For two runs that did not explode (1 and 2) the

Table I. Experimental Conditions for Injecting Sodium into UO2

Run No.	Sodium Temp, °C	Sodium Mass,	Injection Velocity, m/sec	Injection Height, cm above Surface	UO <sub>2</sub> Temp, °C	UO <sub>2</sub> Mass,
1	400	∿5	5.0	1.0	2800**	29.27
2	408	∿5	5.0	1.0	2800**	29.27
3	392	5.7	5.0	1.0	2950	29.27
4	392	∿5	3.5	1.0	2980	31.42
5	395	∿5	*	*	3060	51.06
6	395	∿5	7.5	2.3	3000**	51.06
7	400	∿5	7.5	2.3	2960	49.02
8	400	5.2	7.5	2.3	3090	49.02
9	400	5.1	9.5	2.2	3180	49.59
10	400	3.6	*	*	2900**	51.29
11	400	5.4	*	*	2930	51.29
12	405	6.3	3.0	-1.9	2920	62.08

 $<sup>^{*}</sup>$ Equipment malfunction prevented injection.

 ${\rm UO}_2$  was not molten at the time of injection although in each case the limit of the heating capabilities of the apparatus had been reached. After Run No. 2 the transmission line connecting the induction generator and stepdown transformer (28 ft in length) was modified from parallel, 314-in.-OD copper tubing to parallel, 4-in.-wide copper plates insulated by double 1/8-in. plates of glass. This increased the heating efficiency, and no further difficulties were encountered in melting the  ${\rm UO}_2$ . In Run No. 7 the normal  ${\rm UO}_2$  load of crushed pellets was topped off with a layer of  ${\rm UO}_2$  powder because insufficient pellets were on hand. This powder sintered, forming a crust near the top of the crucible, while the remaining  ${\rm UO}_2$  melted beneath it. The injected sodium was unable to penetrate this crust, and thus there was no contact of sodium and molten  ${\rm UO}_2$ .

The one remaining run that did not produce an explosion (Run No. 4) was a low-velocity, above-surface injection into fully molten  $\rm UO_2$ . A postrun examination of the crucible and  $\rm UO_2$  showed a smooth, conical depression in the  $\rm UO_2$  surface and led to the assumption that the sodium velocity was too low to allow penetration of the molten  $\rm UO_2$ .

<sup>\*\*</sup>Smoke from cooling-oil leak prevented accurate pyrometer readings; the listed temperatures are estimated from heating-power measurements.

Excursions were produced in Runs 3, 6, 8, 9, and 12. Table II lists the experimental data obtained from each of these runs.

Run 6 is included in the table simply because it did produce an explosion, although there was a total instrument failure during the experiment. The camera view was completely obscured by smoke due to an oil leak in the cooling system. Both the force transducer and blast microphone were disabled by sodium contamination of the signal-lead connectors, causing short circuits.

The data in the table are of varying precision due to the circumstances associated with each experiment. There was some time jitter accompanying the start of injection due to the residual magnetism in the latch mechanism. Thus the listed start of injection was preceded by a slow ejection of sodium for a period ranging from 1 to 10 msec, depending on the selected injection rate. Estimates of the total amount of sodium injected during this time range from 0.1 to 0.05 cm<sup>3</sup>, based on test runs with water. The data from the injector had the appearance of a critically damped sine wave superimposed on a linear ramp. The first event in Run No. 9 is the only one in which this time jitter has significance (the effect on delay time would be less than 10% in the other runs). Our best estimate of the true delay time is between 10 and 15 msec. The listed total amount of sodium has been corrected to the best estimate, although again the only run significantly affected is the first event in Run 9.

The total amount of  $\mathrm{UO}_2$  ejected is based on difference weighings of the crucible before and after the experiments, and thus does not reflect the various times at which this ejection took place. An examination of the high-speed motion pictures (see Fig. 2) of the experiments shows that a small amount of the  $\mathrm{UO}_2$  was ejected as small droplets by the violently boiling sodium prior to the explosion, a larger amount was ejected at high velocity by the explosion (see Fig. 2b), followed several milliseconds later by the relatively slow ejection of the remaining  $\mathrm{UO}_2$  (see Fig. 2c), either by the elastic rebound of the apparatus or by sodium vapor generated by the continuing injection stream impacting on the hot crucible.

The period of sodium injection prior to explosion was accompanied by violent boiling of the sodium and the production of copious quantities of sodium vapor, as well as the ejection of many small drops of liquid sodium. This introduced two very serious uncertainties in the data. The first is making the amount of sodium available for interaction at the time of an explosion almost a complete unknown. In Run No. 3, for example, the liquid droplets of sodium escaping from the crucible prior to the explosion were measured from the high-speed films and accounted for 25% of the sodium injected to that time. The amount lost as vapor is completely unknown, but must also be a very significant amount. So it is not unreasonable to expect that the amount of sodium interacting to produce an explosion was less by an order of magnitude than that listed in the table. Indeed as shown in Table III, a listing of the relative efficiencies based on the amount of sodium in two similar explosions differed by a factor of 10, which is probably best explained by the more efficient explosion occurring ~10 msec after the start of injection and thus allowing little time for the loss of sodium.

The second effect of the sodium loss was associated with the production of large quantities of vapor. The shock wave generated by the explosion

Table II. Experimental Data from Sodium Injection into Molten  ${\tt UO}_2$ 

Run No.	Time from Start of Injection to Explosion, msec	Sodium Injected at Time of Explosion, 8	Total UO <sub>2</sub> Ejected,	Peak Reaction Force, 1b	Reaction Impulse, lb-sec	Peak Overpressure, psi	Explosion- Front Velocity, m/sec
3	280	2.5	13.9	400	0.095	6	160
6	a	a	a	a	a	a	a
8	35 & 43	0.56 & 0.62	22.5	45.6 & 22.2	0.0137 & 0.0067	b	44.2 & 15
9	9 & 46.2	$0.14^2 \& 1.37$	42.25	399 & 477	0.11 & 0.107	4.91	157 & c
12	378	0.84	36.72	213	0.08	ь	94.53

<sup>&</sup>lt;sup>a</sup>Subjectively this produced the largest explosion of the series; however, total instrumentation failure occurred.

Table III. Energy and Efficiency of Sodium-vapor Explosions

		gy, J	Efficiency, %				
<u> </u>		asis			Basis		
Run No.	Total Mass Ejected	Front Velocity	Particle Velocity	UO <sub>2</sub> Thermal	Thermodynamic		
3	5.4	33.6		0.08	1.2		
6				A k Italia			
8	0.17	1.57		0.002	0.07		
9	0.72	38.7	155	0.05	6.8		
12	2.3	15.7	23.9	0.02	0.77		

bBlast-microphone failure.

<sup>&</sup>lt;sup>C</sup>Second event obscured from first event.

Thermal shock from first event overwhelmed second event.

Best estimate (see text).

Average of several measurements.







Fig. 2. Selected Frames from Motion Picture of Sodium-UO<sub>2</sub> Interaction.

(2a) ANL Neg. No. 900-76-30.

(2b) ANL Neg. No. 900-76-29.

(2c) ANL Neg. No. 900-76-31.

passing through the vapor produced a condensation cloud which very effectively obscured the camera view of events following the explosion. In some runs, low-density areas or "windows" in the cloud allowed a partial view of the following events while others were nearly completely obscured.

The force measurements were reliable for all but the one run (6) already mentioned; however, the blast-microphone data were much less reliable. Of necessity, the diaphragm of the microphone was exposed to the atmosphere surrounding the crucible and thus was subject to thermal shocks from moving incandescent parts of the apparatus initially, and later to ejected sodium and  $\rm UO_2$ . Although there was little possibility of confusing thermal and blast effects, the thermal effects were of much greater magnitude and often overwhelmed the recording system.

The primary purpose of these experiments was to investigate the existence of vapor explosions with reactor materials. However since the direct data from the experiments are of little use except, however importantly, to prove that reactor materials can explode, further uncertainties had to be introduced in manipulating these data into a more useful form.

## V. DATA REDUCTION

The force history recorded during the experiment could be integrated with respect to time to provide the reaction impulse produced on the crucible by the explosion. These impulse data appeared relatively good, and a test made by dropping a steel ball onto a plate clamped on the crucible helped to confirm the reliability of the data. However, the determination of the mechanical energy produced by the explosion required an additional piece of data, either the mass of the material expulsed by the explosion or the velocity. A direct determination of the mass expelled at the time of the explosion was impossible from the recorded data because the film record of the event showed only an incandescent expansion front (a mixture of molten UO2 particles and UO2 and sodium vapor, with no measurable internal detail). difference measurement of the total amount of UO2 expelled during the experiment was obviously unusable both because the film record showed a large amount of UO2 expelled after the explosion and because calculations based on this mass gave velocities much lower than the minimum observed. The measurement of the velocity of the expelled mass, although possible, is degraded by several uncertainties. First, the measurement determines the velocity of the expansion front, and the relationship between this and the mass average velocity at any time is unknown. However, it certainly provides an upper limit. A second uncertainty is the reliability of the measurement itself. Due to the poor viewing conditions in the furnace chamber it was never certain whether one was seeing the movement of the expansion front or the shifting of the windows in the condensation cloud. In Run No. 9, for instance, the expulsion velocity measured by film observation of the expansion front was 15 m/sec, whereas that calculated from backtracking of trajectories of UO2 particles penetrating the condensation cloud was 630 m/sec.

Table III lists the mechanical energies developed in each of the explosions as determined by different methods of calculating the expelled mass. In addition, the relative conversion efficiency of thermal to mechanical energy based on the energy determined from the expansion-front

velocity is listed in two forms. The first is based on the total thermal energy above  $20^{\circ}\text{C}$  in the  $\text{UO}_2$ , and the second is based on the maximum thermodynamic conversion possible, i.e., a constant-volume heating of the sodium to the  $\text{UO}_2$  temperature followed by an isothermal expansion to the void volume of the crucible (a modified Hicks and Menzies acalculation). As mentioned earlier, the mass of sodium used for this calculation was taken as the total amount injected at the time of the explosion which may be an order of magnitude greater than that actually involved.

The wide spread in the results of these experiments, i.e., a factor of 30 in the delay time, 20 in the energy, and 100 in the thermodynamic efficiency, combined with the limited number of successful runs (4) make it very difficult to reach any concrete conclusions about  $\rm UO_2/sodium\mbox{-}vapor$  explosions. However, this work does demonstrate conclusively that vapor explosions are possible with reactor materials. For possible interpretation of the cause of these explosions see Refs. 4 and 5.

### REFERENCES

- 1. D. R. Armstrong, F. J. Testa, and D. C. Rardin, Interaction of Sodium with Molten  $UO_2$  and Stainless Steel Using a Dropping Mode of Contact, ANL-7890 (December 1971).
- 2. Reactor Development Progress Report, ANL-RDP-2, p. 8.31 (February 1972).
- 3. G. P. Hicks and D. C. Menzies, Theoretical Studies on the Fast Reactor Maximum Accident, ANL-7120, pp. 654-685 (October 1965).
- 4. H. K. Fauske, The Role of Nucleation in Vapor Explosions, Trans. Am. Nucl. Soc., 15(2), 813 (November 1972).
- 5. R. P. Anderson and D. R. Armstrong, Comparison between Vapor Explosion Models and Recent Experimental Results, AIChE Symposium Series No. 138, 70, pp. 31-47 (1974).

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